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(11) EP 0 966 182 A1

(12)

#### **EUROPEAN PATENT APPLICATION**

(43) Date of publication: 22.12.1999 Bulletin 1999/51

(51) Int. Cl.<sup>6</sup>: **H05B 33/10**, H01L 51/20

(21) Application number: 98306730.7

(22) Date of filing: 21.08.1998

(84) Designated Contracting States:

AT BE CH CY DE DK ES FI FR GB GR IE IT LI LU

MC NL PT SE

Designated Extension States:

AL LT LV MK RO SI

(30) Priority: 17.06.1998 KR 9822756

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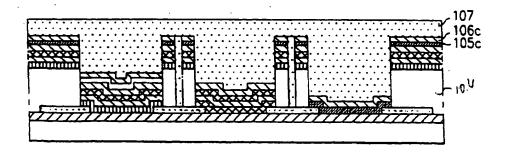
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### (54) Method of fabricating organic electroluminescent display panel

(57) Disclosed is a method of fabricating an organic EL display panel wherein pixelation can be performed without resorting to the use of a shadow mask and also without active EL elements being exposed to solvents from photoresist or developing and stripping solutions. The fabrication scheme comprises the steps of: (1) forming a first electrode layer and an insulating layer, in succession, on a transparent substrate; (2) removing said insulating layer in the predefined region by a single or a plurality of laser beams; (3) forming organic function layers and a second electrode layer, in succession, on the predefined surface inclusive of the insulating layer; and (4) repeating the steps of (2) and (3) once or more.

FIG.7H



#### Description

#### BACKGROUND OF THE INVENTION

#### 5 Field of the Invention

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[0001] The present invention relates to a novel method of fabricating an organic electroluminescent (hereinafter referred to as EL) display panel comprising organic EL elements which emit light when electric charges are injected into them.

#### Discussion of the Background Art

[0002] The technology of organic EL devices, also called organic light emitting diodes (LEDs), has been rapidly advancing, and several prototype modules have been successfully demonstrated at exhibitions. Organic EL devices are extremely thin, matrix-addressable and operable at a relatively low voltage, typically less than 15 volts. Furthermore, they have additional features suitable for next generation flat panel displays (FPDs) such as, among other things, little dependence on viewing angle and good device-formability on flexible substrates. A major drawback of liquid crystal display, currently most well-known display of choice, is that most of them require bright backlighting, which can be easily eliminated by the use of an organic EL display.

[0003] Organic LEDs differ fundamentally from conventional inorganic LEDs. While the charge transfer in inorganics is band-like in nature and the electron-hole recombination results in the interband emission of light, organic films are generally characterized by the low-mobility activated hopping transport and the emission is excitonic. Organic EL devices are also substantially different from conventional inorganic EL devices, especially in that organic EL devices are operable at low DC voltages.

[0004] A substantial amount of research has been directed towards the efficiency improvement and color control of organic LEDs. The efficiency of organic EL devices has now been demonstrated to be certainly adequate for many commercial applications. Moreover, color control is probably not limiting for most potential applications. In light of this, we believe that the outlook for commercial applications is excellent for organic EL devices. The performance of the organic EL devices is quite satisfactory for many applications. It is valuable to think in terms of specific products and manufacturing techniques for the commercialization of organic EL devices. Consideration of the specific applications leads us to believe that more work on manufacturability, uniformity, reliability, and systems issues is required to commercialize organic EL.

[0005] The simplest way to drive an organic EL panel is to have organic function layers sandwiched between two sets of orthogonal electrodes, i.e. rows and columns (Fig. 1). In this passive addressing scheme, the EL element serves both the display and switching functions. The diode-like nonlinear current-voltage characteristic of the organic EL element should, in principle, permit a high degree of multiplexing in this mode of addressing.

[0006] Pixelation or patterning, especially of electroluminescent and second electrode materials, is one of the key issues to be resolved before the commercialization of organic EL devices. The use of many conventional pixelation techniques is precluded due to the nature of organic materials which are extremely vulnerable to the attack by most solvents.

[0007] The simplest patterning method is probably to use a shadow mask. As shown in Figs. 1 and 2, the pixelation of organic EL display panel can be accomplished by depositing second electrode material(s) 4 through the openings of a shadow mask 5 onto organic function layers 3 which are, in turn, laminated on a plurality of first electrode stripes 2. Said first electrode stripes 2 are generally formed by patterning a layer of indium tin oxide (ITO) deposited on a transparent, insulating substrate 1.

[0008] The pixelation method using a shadow mask becomes less efficient as the display resolution becomes finer. One possible solution for a monochrome display is to separate adjacent pixels using electrically insulating ramparts 6 as suggested in U.S. Patent No. 5,701,055 (Fig. 3).

[0009] As disclosed in JP Laid Open Patent No. H8-315981 (Figs. 4a ~ 4d), the use of additional shadow masks may be required for the construction of a multi or full color display: (1) putting a shadow mask with a plurality of openings (5-1, 5-2 or 5-3) onto top surfaces of the ramparts 6, and aligning each of said openings to be placed over the gap between corresponding ramparts; (2) depositing organic EL medium layers of red (R), green (G) and blue (B) (4-1, 4-2 and 4-3), one by one, through said openings; and (3) forming at least one second electrode layer 3 on said ramparts and said organic function layers

[0010] The above method may work fine for a display of moderate resolution and size. As the display size increases and the pitch decreases, however, said method reveals limitations: difficulties in making a shadow mask itself and additionally in aligning said shadow mask with respect to a substrate. The need to make a pixelation without resorting to shadow mask(s) has been, in part, answered by U.S. Patent No. 5,693,962 and JP Laid Open Patent No. H9-293589.

[0011] Figs. 5a ~ 5c illustrate the fabrication steps of a full color panel as suggested in U.S. Patent No. 5,693,962. Fig. 5a describes the formation process of a first sub-pixel: (1) patterning a layer of organic or inorganic conductor deposited on a transparent substrate 11 by conventional lithographic techniques to form a plurality of parallel conductive stripes for the first electrode 12; (2) depositing a layer of dielectric medium 13 on top of said conductive stripes 12 and the exposed portions of substrate 11; (3) spin-coating of a photoresist (PR) layer 14-1 and patterning said dielectric medium 13 by a dry or wet etching technique; (4) laminating an electroluminescent medium 5-1; (5) forming a capping layer 17-1 of the sub-pixel by depositing an air-stable metal on top of said electroluminescent medium; and (6) removing the PR layer 14-1 and those on said PR layer by lift-off. Acetone or a stripping solution is often used for lift-off. It is important to note that said electroluminescent medium can be exposed, through said capping layer, to said solvents during said lift-off process. The device degradation due to said exposure is not surprising, given the relatively poor adhesion between an organic material and a metal used as a capping layer. As shown in Figs. 5b and 5c, second and third sub-pixels can be fabricated by the same token.

[0012] Figs. 6a ~ 6j describe the processes of forming R, G, B sub-pixels as published in Japanese Laid Open Patent No. H9-293589: (1) forming a first electrode layer 22 on a transparent substrate 21, followed by the lamination of an electroluminescent medium (R) 23, a second electrode 24, and a protection layer 25; (2) spin-casting of photoresist (PR) 26 and subsequent patterning of red sub-pixels; and (3) repeating the above steps to form green and blue sub-pixels. During the above processing steps, active display elements are substantially exposed to PR, which is detrimental to device performance because of solvents remaining in PR. For rough evaluation of adverse effects of photoresist, it is necessary to understand a typical photolithographic process: spin-casting of a photoresist solution on the substrate, soft-baking and exposure to an ultraviolet ray, developing, and hard-baking. In the background art under discussion, the panel is thereafter supposed to be subjected to etching and then PR stripping which again needs stripping solution(s). During spin-casting, active EL elements are already exposed to solvents from the photoresist in a liquid phase.

[0013] In short, background methods described above have a serious drawback. Active EL elements are inevitably

exposed to various solvents from photoresist or developing and stripping solutions. Said solvents are extremely harmful to electroluminescent medium, and adversely affects the device performance.

#### SUMMARY OF THE INVENTION

[0014] Accordingly, the present invention is directed to a novel method of fabricating an organic electroluminescent display panel that addresses one or more of the problems due to limitations and disadvantages of the related art.

[0015] An object of the present invention is to provide a method of fabricating an organic EL display panel wherein pixelation can be performed without resorting to the use of a shadow mask and also without active EL elements being exposed to solvents from photoresist or developing and stripping solutions.

[0016] Additional features and advantages of the invention will be set forth in the description which follows, and in part will be apparent from the description, or may be learned by practice of the invention. The objectives and other advantages of the invention will be realized and attained by the structure particularly pointed out in the written description and claims hereof as well as the appended drawings.

[0017] In one aspect the present invention provides a method of fabricating an organic electroluminescent (EL) display panel having organic function layers including at least one organic EL medium layer laminated between first and second electrodes comprises the step of pixelation by a single or a plurality of laser beams.

[0018] In another aspect of the present invention, there is provided a method of fabricating an organic EL display panel, comprising the steps of: (1) forming a first electrode layer, organic function layers including at least one organic EL medium layer, and a second electrode layer in succession on a transparent substrate; and (2) removing, selectively, said second electrode layer and organic function layers in the predefined region by a single or a plurality of laser beams.

- [0019] In additional aspect of the present invention, there is provided a method of fabricating an organic EL display panel, comprising the steps of: (1) forming a first electrode layer and an insulating layer in succession on a transparent substrate; (2) removing said insulating layer in the predefined region by a single or a plurality of laser beams; (3) forming organic function layers and a second electrode layer, in succession, on the predefined surface inclusive of the insulating layer; and (4) repeating the steps of (2) and (3) at least once.
- [0020] In further aspect of the present invention, there is provided a method of fabricating an organic EL display panel, comprising the steps of: (1) forming a first electrode layer and an insulating layer in succession on a transparent substrate; (2) removing said insulating film in the predefined region by a single or a plurality of laser beams; (3) forming organic function layers, a second electrode layer and a protection layer, in succession, on the predefined surface inclusive of the insulating layer; and (4) repeating the steps of (2) and (3) once or more.
- [0021] It is to be understood that both the foregoing general description and the following detailed description are exemplary and explanatory and are intended to provide further explanation of the invention as claimed.

#### BRIEF DESCRIPTION OF THE DRAWINGS

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[0022] The accompanying drawings, which are included to provide further understanding of the invention and are incorporated in and constitute a part of this specification, illustrate embodiments of the invention and together with the description serve to explain the principles of the invention.

[0023] In the drawings:

- Fig. 1 illustrates a plan view of a typical passive addressing organic EL display panel;
- 10 Fig. 2 is a cross-sectional view showing the process of patterning the second electrode using a shadow mask;
  - Fig. 3 is a sectional view illustrating the process of pixelation using insulating ramparts;
  - Figs. 4a~4d, 5a~5c and 6a~6j illustrate sectional views showing the fabricating steps of background methods;
  - Figs. 7a~7h illustrate sections showing the steps of fabricating an organic EL display panel in accordance with the first embodiment of the present invention;
- Figs. 8a~8k illustrate sections showing the steps of fabricating an organic EL display panel in accordance with the second embodiment of the present invention;
  - Fig. 9 illustrates an exemplary laser-beam etching system in accordance with an embodiment of the present invention; and.
- 25 Fig. 10 illustrates an exemplary process of the laser-beam etching using a mask in accordance with an embodiment of the present invention.

#### DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENT

- 30 [0024] Reference will now be made in detail to the preferred embodiments of the present invention, examples of which are illustrated in the accompanying drawings. For the fabrication of an organic EL display panel with a plurality of emitting portions laminated between a plurality of first and second electrodes, a pixelation method is utilized characterized by employing a laser-beam etching method, by not using a shadow mask for pixelation, and also by preventing active EL elements from being exposed to various solvents from photoresist or developing and stripping solutions. Though the organic EL display panel has a plurality of R(red), G(Green) and B(Blue) sub-pixels, for clarity, only a single set of R, G and B sub-pixels is illustrated in the accompanying drawings.
  - [0025] Figs. 7a~7h illustrate sectional views showing the steps of fabricating an organic EL display panel in accordance with the first embodiment of the present invention. Referring to Fig. 7a, the fabrication processes start with forming the stripes of first electrodes 102 by photolithographically patterning a transparent layer of typically indium tin oxide (ITO) deposited on a transparent substrate 101, and forming the stripes of an electrically insulating buffer layer 103 in orthogonal relationship to said stripes of first electrodes 102. Said buffer layer 103, being not luminescent, serves for the reduction of leakage current by lowering the chance of first electrodes 102 to be microscopically short-circuited to second electrodes that are to be formed in a later step. Said buffer layer 103 can be formed by depositing preferably an inorganic compound such as silicon oxide or silicon nitride or a polymeric material such as polyimide by vapor deposition, e-beam evaporation, RF sputtering, chemical vapor deposition (CVD), spin coating, dipping, Dr. blade method, electro- or electroless plating, or screen printing method.
  - [0026] Then, as shown in Fig. 7b, an insulating layer 104 is formed on the predefined surface inclusive of said buffer layer 103, and a laser-beam etching is performed to remove a portion of said insulating layer 104 wherein red emitting sub-pixels are to be formed in the next step. The material for said insulating layer 104 needs to have a considerable absorption cross-section at the wavelength of the laser beam used and additionally a good film-forming characteristic. Said material is preferably polymeric, for example photoresist, and is preferably formed to a thickness of 0.1 ~ 100 µm. [0027] The most distinctive difference between the background art and the embodiment lies in the fact that (1) the process involving a photoresist is performed before the lamination of any electroluminescent medium, and (2) therefore, the photoresist used can be hard-baked at a high enough temperature for a long enough time, ensuring complete elimination of any residual solvents inside the PR film. If an EL medium is subjected to that harsh a condition, it will degrade very quickly. Thus, the photoresist used in the background art can not be completely baked. To the contrary, the photoresist used here does not pose any adverse effects onto the device performance.
  - [0028] In the next step, the part of the ITO-coated substrate wherein sub-pixels are to be formed is optionally treated

with oxygen plasma or UV/ozone. As shown in Fig. 7c, next, the first organic function layers 105a (for example, red light emitting) and a second electrode layer 106a are deposited, in succession, on the predefined surface inclusive of the insulating film 104. A typical example of organic function layers 105a is: (1) a buffer layer of copper phthalocyanine (CuPc) typically 10nm ~ 20nm thick, (2) a hole transporting layer of N,N'-diphenyl-N,N'-bis(3-methylphenyl)-(1,1'-biphenyl)-4,4'-diamine (TPD) typically 30nm ~ 50nm thick, and (3) an emitting layer of tris(8-hydroxy-quinolate)aluminum (Alq<sub>3</sub>) 40nm ~ 60nm thick, doped with DCM 2 for the red emission. The second electrode 106a is typically formed of any one selected from Al, Ca, Mg:Ag, and Al:Li.

[0029] Next (Fig. 7d), another portion of said insulating layer 104 is etched out with a laser beam. Following an optional surface treatment, the second organic function layers 105b (for example, green light emitting) and a second electrode layer 106b are deposited (Fig. 7e). A typical green light emitting EL medium is coumarin 6 doped Alg<sub>3</sub>.

[0030] Figs. 7f and 7g illustrate the repetition of a sub-pixel formation for the third color, blue in this instance. BAlq<sub>3</sub> doped with perylene is an example of blue emitting EL medium. To ensure the electrical insulation between two adjacent sub-pixels, it is appropriate to cut into two parts, using a laser-beam, the organic function layers and second electrode layers formed on top of an insulating layer 104 between two adjacent sub-pixels (Fig. 7h). Next, the panel is covered with a protection layer 107 to prevent primarily moisture from penetrating into active EL elements, and is consequently subjected to an encapsulation process. Said protection layer 107 may contain one or a mixture of moisture absorbing incredients in it. The formation of a protection layer may be performed prior to the laser scribing.

[0031] Figs. 8a~8k illustrate sections showing the steps of fabricating an organic EL display panel in accordance with the second embodiment of the present invention, which is exactly the same as the first embodiment except that a protection film is coated on top of a sub-pixel every time said sub-pixel is formed. The first and second sub-pixels are coated with protection layers 107a and 107b before the formation of the second and third sub-pixels, respectively (Figs. 8d & 8g), and a protection layer 107c is additionally coated after the formation of the third sub-pixel (Fig. 8j). Since said protection layers 107a and 107b can help to shield the first and second sub-pixels, respectively, from moisture and solvents during the subsequent process steps, the long-term stability should improve and, furthermore, more freedom in the process design can be gained. Said inner protection layers 107a, 107b and 107c remain under the outermost protection layer 107d even after the completion of device fabrication. Therefore, it is highly recommended for said inner protection layers to contain one or a mixture of moisture absorbing ingredients in them.

[0032] The beauty of this lies in the simplicity of a sequence of sub-pixel formation steps, i.e the repetition of the laser-beam etching and the formation of EL elements. The details of laser-beam etching techniques and process requirements are as follows.

[0033] In order to prevent degradation of active display elements during the etching process, the panel under process needs to be located either in vacuum or in a dry or inert atmosphere while the laser beam generator may be operated in the atmosphere. The laser beam is guided into a vacuum chamber or a glove box filled with an inert gas through a window having a limited absorption of the laser beam. Depending on the system design, either the laser beam is scanned (Fig. 9), or the moving stage with a panel mounted on it moves as programmed, generally the former method being less suitable for applications requiring such an accurate position control as in the present invention. Said moving stage may have encoders or sensors attached for a feedback control. It has been found useful that optical and electrical properties of ITO can be used for the feedback control of positioning.

[0034] A laser beam with a proper wavelength should be selected considering physical and chemical properties of the materials to be etched, of which the most important factor is the absorption cross-section as a function of wavelength. The lasers much used for etching metals and organic materials are a frequency doubled Nd:YAG laser with a green emission and an excimer laser in ultraviolet. In the present invention, the laser is preferably operated in a pulse mode rather than a continuous mode which may cause a thermal damage to the device. The laser power and pulse repetition rate should be optimized to ablate the second electrode layer and organic function layers, but not to damage the first electrode. A list of lasers usable in the present invention is as follows:

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laser	gas	wavelength(	remark
Nd:YAG		1.06	
•		0.53	Frequency
Er:YAG		2.9	
Ho:YAG		1.9	
Excimer laser	F <sub>2</sub>	0.16	
•	ArF	0.19	

#### (continued)

laser	gas	wavelength(	remark	
	KrF 0.25			
•	XeCI	0.31		
	XeF	0.35		
Ar ion laser	Ar	0.52		
N <sub>2</sub> laser	N <sub>2</sub>	0.34		

[0035] Depending on the process design, the laser beam may be directed into the panel from the first electrode side or from the second electrode side. ITO, a typical first electrode material, apparently does not have a large absorption cross-section at 0.53 μm, and thus a frequency doubled Nd:YAG laser may be irradiated from the ITO side to etch organic function layers and the second electrode. Etching rates of certain materials may be enhanced if a single or a plurality of appropriate reactive gases are introduced together with the laser beam. The shape and size of a laser beam spot may be adjusted as necessary. In addition, a plurality of sub-pixels may be etched simultaneously with the use of a properly designed etching mask, as shown in Fig. 10. Irradiating a large area at a time would require a laser system with a large beam spot and a high power.

[0036] The method of fabricating an organic EL display panel has the following advantages.

[0037] Primarily, the production yield is significantly raised because a simple and fast laser processing method is employed and also because a shadow mask, difficult to make and even more tricky to accurately align with respect to a substrate, is not used for the fabrication. Furthermore, the long-term stability is much improved by minimizing the exposure of active EL elements to detrimental solvents and moisture.

[0038] It will be apparent to those skilled in the art that various modifications and variations can be made in the method of fabricating an organic EL display panel of the present invention without departing from the scope of the invention. Thus, it is intended that the present invention cover the modifications and variations of this invention provided they come within the scope of the appended claims and their equivalents.

#### 30 Claims

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- 1. A method of fabricating an organic EL display panel, comprising the steps of:
  - (1) forming a first electrode layer, organic function layers including at least one organic EL medium layer, and a second electrode layer in succession on a transparent substrate; and
  - (2) removing, selectively, said second electrode layer and organic function layers in the predefined region by a single or a plurality of laser beams.
- 2. A method as claimed in claim 1, wherein said laser beam is operated in a pulse mode.
- A method as claimed in claim 1 or claim 2, wherein said laser beam is directed into the panel under process from either side of the transparent substrate.
- 4. A method as claimed in any of the above claims, wherein at least one reactive gas is flown onto the surface of said display panel under process along with the laser-beam irradiation, in the step (2).
  - 5. A method as claimed in any of the above claims, wherein a mask with the predefined pattern is provided between a laser source and the display panel under process, in the step (2).
- 6. A method as claimed in any of the above claims, wherein a protection layer is formed on the surface of the pixelated substrate, after the step (2).
  - 7. A method of fabricating an organic EL display panel, comprising the steps of:
    - (1) forming a first electrode and an insulating layer, in succession, on a transparent substrate;
    - (2) removing, selectively, said insulating layer in the predefined region by a single or a plurality of laser beams;
    - (3) forming organic function layers including at least one organic EL medium layer and a second electrode layer, in succession, on the predefined surface inclusive of said insulating layer; and,

- (4) repeating the steps of (2) and (3) once or more.
- 8. A method as claimed in claim 7, wherein step (3) comprises:
- forming organic function layers including at least said one organic EL medium layer, said second electrode layer, and a protection layer in succession on the predefined surface inclusive of said insulating layer.
  - 9. A method as claimed in claim 7 or claim 8, further comprising the step of cutting into two parts, using a single or a plurality of laser beams, the organic function layers and second electrode layer formed on top of said insulating layer between two adjacent sub-pixels, after the step (4).
  - A method as claimed in any of claims 7 to 9, wherein a buffer layer is provided between the insulating layer and the first electrode.
- 15. 11. A method as claimed in claim 10, wherein said buffer layer is formed of an inorganic or organic material with a good insulating property.
  - 12. A method as claimed in any of claims 7 to 11, wherein said insulating layer is formed of a single or a plurality of polymeric materials.
  - 13. A method as claimed in any of claims 7 to 12, wherein said insulating layer is subjected to a process to eliminate solvents and moisture, after the step (1).
  - 14. A method as claimed in any of claims 7 to 13, wherein said insulating layer has a thickness of 0.1 μm~ 100 μm.
  - 15. A method as claimed in any of claims 7 to 14, wherein a protection layer is formed on the predefined surface of the pixelated substrate, after the step (4).
- 16. A method as claimed in any of the above claims wherein at least one protection layer contains one or a mixture of moisture absorbing ingredients.
  - 17. A method as claimed in any of the above claims, wherein the step (2) is conducted in vacuum or in a dry or inert atmosphere.
- 18. A method of manufacturing an organic electroluminescent (EL) display panel having organic function layers including at least one organic EL medium layer laminated between first and second electrodes comprises the step of pixelation using a single or a plurality of laser beams.
  - 19. An organic electroluminescent display panel manufactured by the method as claimed in any of claims 1 to 18.

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FIG.1 Background Art

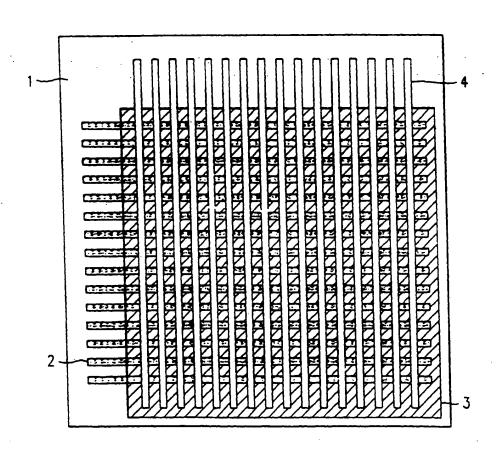


FIG.2 Background Art

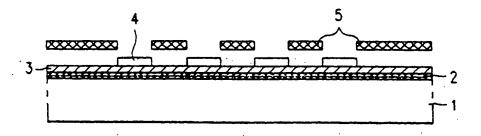


FIG.3 Background Art

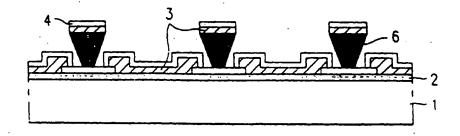


FIG.4A Background Art

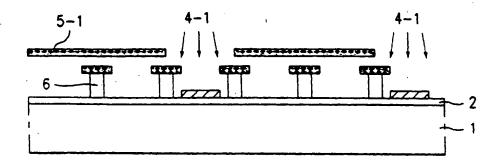


FIG.4B Background Art

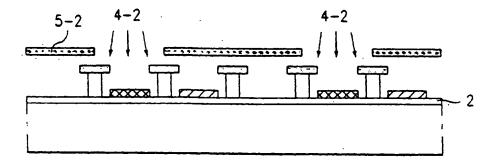


FIG.4C Background Art

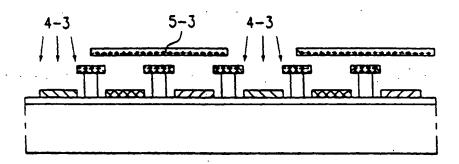


FIG.4D Background Art

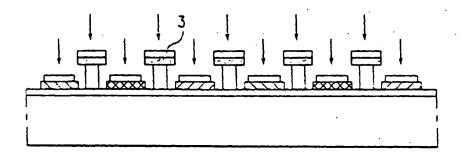


FIG.5A Background Art

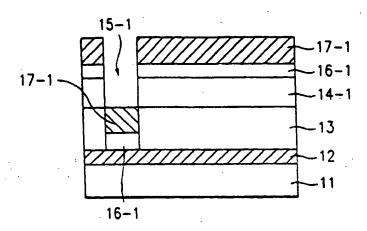


FIG.5B Background Art

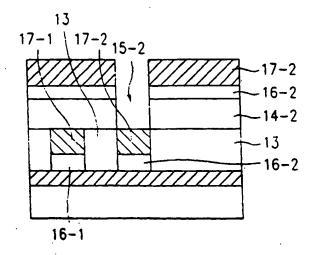


FIG.5C Background Art

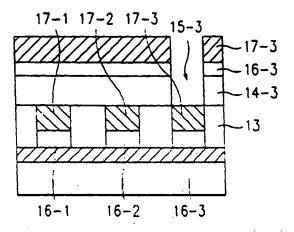


FIG.6A Background Art

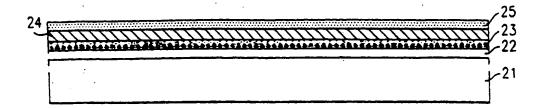


FIG.6B Background Art

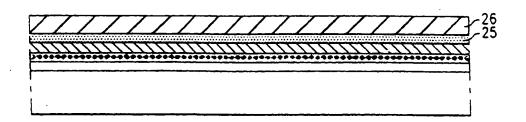


FIG.6C Background Art

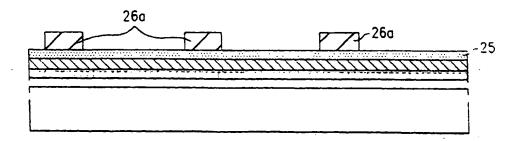


FIG.6D Background Art

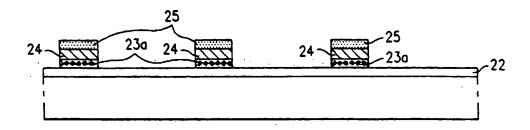


FIG.6E Background Art

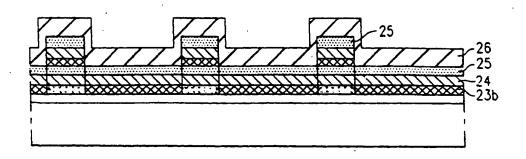


FIG.6F Background Art

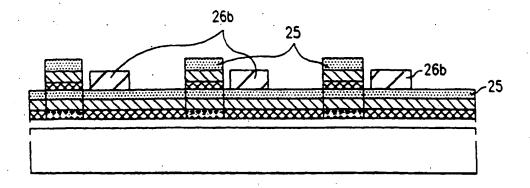


FIG.6G Background Art

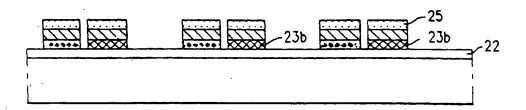


FIG.6H Background Art

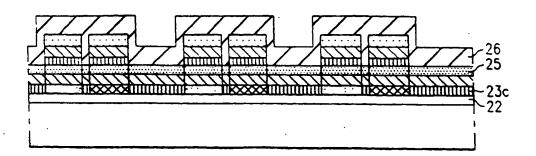


FIG.61 Background Art

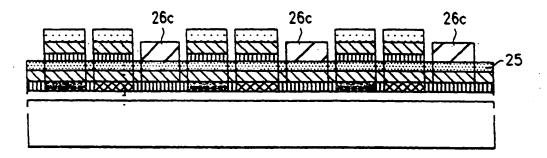
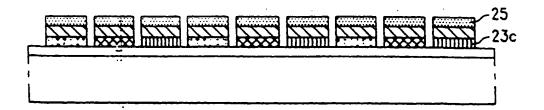


FIG.6J Background Art



# FIG.7A

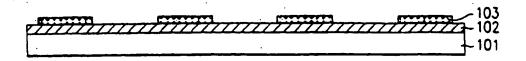


FIG.7B

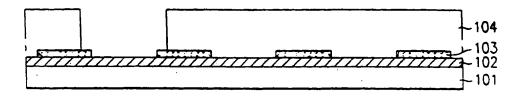


FIG.7C

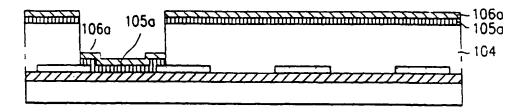


FIG.7D

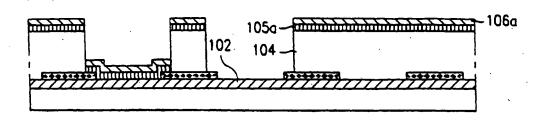


FIG.7E

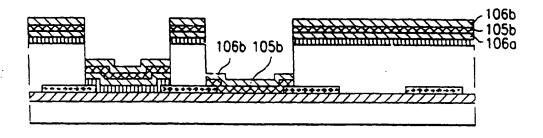


FIG.7F

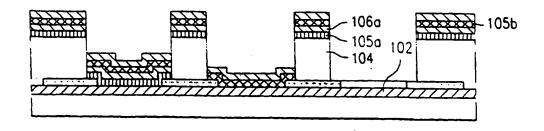


FIG.7G

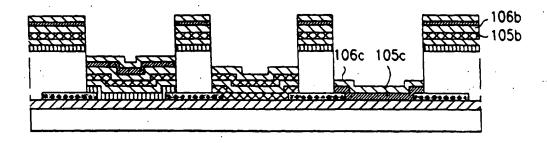
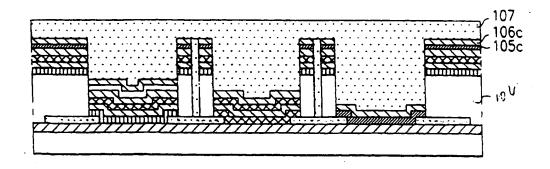


FIG.7H



# FIG.8A

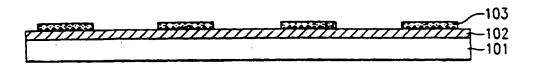


FIG.8B

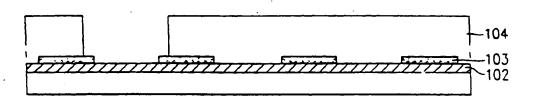


FIG.8C

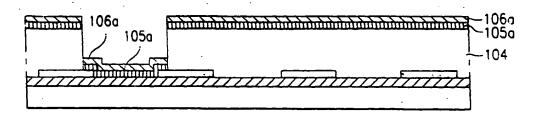


FIG.8D

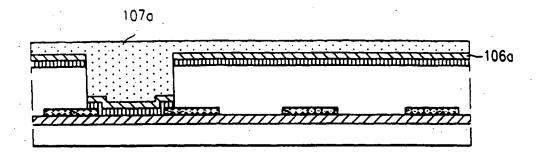


FIG.8E

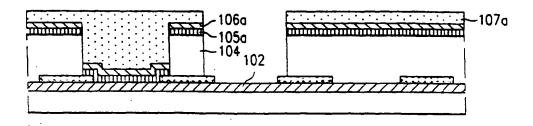


FIG.8F

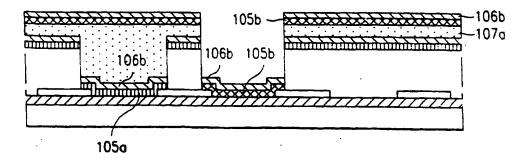


FIG.8G

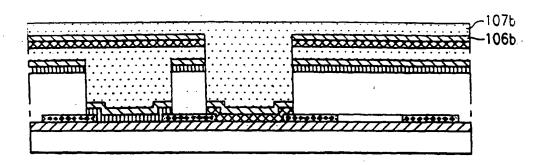


FIG.8H

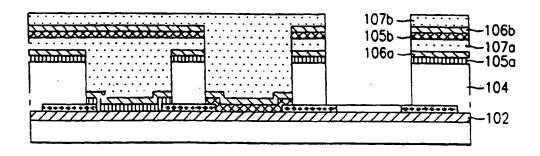


FIG.81

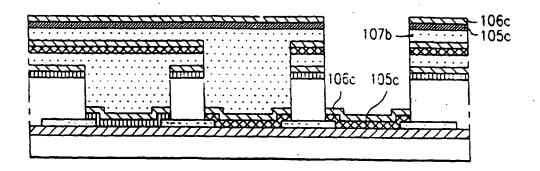


FIG.8J

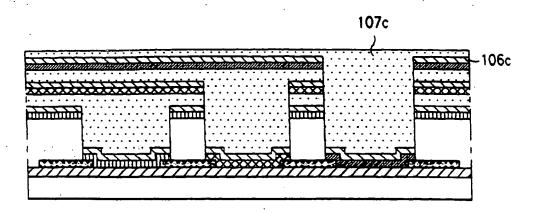


FIG.8K

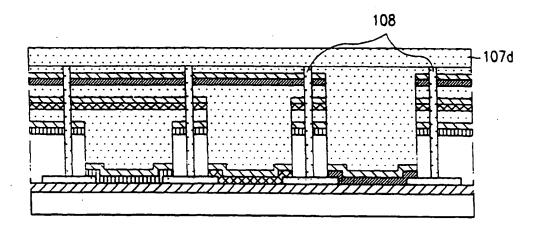


FIG.9

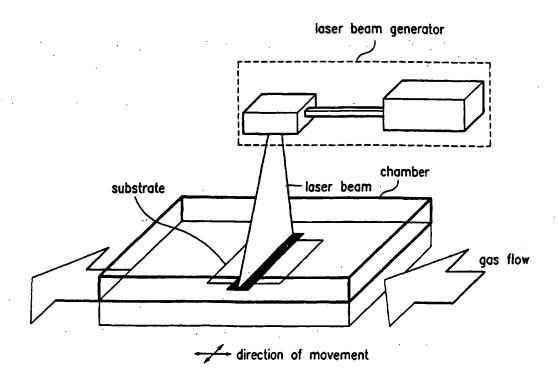
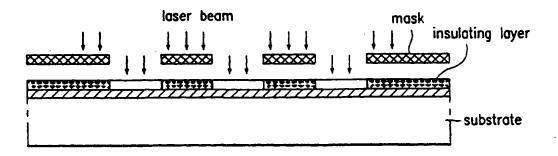


FIG.10





# **EUROPEAN SEARCH REPORT**

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